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LUMINESCENCE STUDY OF NON-EQUILIBRIUM EFFECTS IN LASER GENERATED PLASMA

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Abstract - The properties of surface generated electron-hole plasma in Silicon are studied using luminescence spectroscopy. By detailed line shape analysis we determine plasma densities up to \(2 \times 10^{19} \text{cm}^{-3}\) for 300K and (ns-)pulse energies of 0.08 J/cm². We demonstrate that drift effects are essential for the description of the luminescence. Typical plasma drift velocities are about \(4 \times 10^{6} \text{cm/s}\).

For the description of pulsed laser annealing (PLA) two controversial models have been used. The supporters of the "Thermal Model" generally assume that the excitation energy is transferred immediately to the lattice/1,2/. In consequence the energy transport by electron-hole pairs is usually neglected. In contrast a "Nonthermal Model" has been proposed/3/ which predicts a significant reduction of the melting temperature by a plasma with a density of the order of \(10^{21} \text{cm}^{-3}\). If those densities are to be achieved in high excitation experiments one has to assume that recombination (Auger effect) and diffusion are strongly inhibited/4/.

In order to verify one of these models a wealth of experimental results including reflection, transmission and Raman data has been published/5-7/. From the experimental data lattice and plasma temperatures as well as plasma densities are estimated using simple classical models. However, at the high densities obtained in PLA the carrier system approaches the quantum limit and many body effects as well as non-equilibrium features are expected to become important.

We have used luminescence experiments of highly excited Silicon to study many body and non-equilibrium transport properties of the plasma. Emission spectroscopy of Si is particularly suited for this investigation for the following reasons:
- High excitation experiments at liquid Helium temperature show the formation of electron-hole droplets (EHD). The spectral shape and the band renormalisation by many body effects are in quantitative agreement with theory/8/.
- Above the critical point for a droplet condensation (23K) a non equilibrium electron-hole plasma (EHP) is observed/9/.

The subject of the present study is to apply the methods of measurement and analysis developed in low temperature experiments to PLA conditions. The experimental setup is described with special emphasis on the laser power limit imposed by the onset of surface damages. We describe the line shape analysis and we demonstrate that the plasma expansion alters significantly the emission spectra. Our analysis yields unexpected high plasma drift velocities and implies that plasma expansion effects must not be neglected in any quantitative theory of PLA.
I - Experiment

The samples of very high purity Si (specific resistance $\rho = 8000 \, \Omega \text{cm}$) are situated in a temperature controlled dewar (temperature range 1.8K - 400K) and excited by a frequency doubled Nd-YAG laser with a pulse length of 65 ns.

The sample surface can be modified by strong laser pulses. For luminescence measurements however, the sample must not be altered. Therefore the highest laser power that can be used is close below the threshold for surface modification. Above an energy density of $2.0 \, \text{J/cm}^2$ (one pulse) the surface is destroyed. The threshold for annealing effects identified by the occurrence of c-Si regions in a-Si has been determined as $0.32 \, \text{J/cm}^2$. In contrast to the single pulse experiments surface change can be observed at lower laser pulse energies by using multiple pulses. Hereby the surface damage consists of rings of ripples at the edge of the excitation spot and this is quite different to the patterns seen in single pulse experiments. The threshold for a multiple pulse surface change is determined as $0.09 \, \text{J/cm}^2$.

The luminescence radiation was detected by a selected infrared sensitive Si photo multiplier and processed by a photon counting system. The detection gate had a length of 20 ns and was situated at the maximum of the laser pulse.

II - Luminescence Analysis

Fig. 1 shows a series of plasma luminescence spectra recorded at different bath temperatures at the excitation pulse energy of $0.08 \, \text{J/cm}^2$. With increasing temperature the emission line width increases whereas the energetic position of the intensity maximum is unchanged.

From an analysis of the experimental luminescence spectra with calculated emission bands average values of the plasma density and temperature as well as the band renormalization can be determined. In the following a line shape calculation used successfully for the description of the low temperature plasmas is outlined briefly with special emphasis on modifications necessary for the characterization of the non-equilibrium plasmas under PLA conditions.

To a first approximation the emission line shape of a band-to-band transition in an indirect gap semiconductor is described by the convolution of the densities of occupied states in the conduction and the valence bands. The densities of electrons and holes in the bands are characterized by quasi-Fermi levels. In the case of a moderately degenerate plasma the sum of electron and hole quasi-Fermi levels determines the emission line width whereas the carrier temperature can be obtained from the high energy tail.\cite{10}. Note that due to the wide range of temperature studied the evaluation has to include the explicit temperature dependence of the effective masses. The effective masses of electrons and holes at 300K are larger by about 10% and 30% respectively than their 0K values\cite{11}.
III - Band Gap Renormalization

The energetic position of the low energy edge of the plasma luminescence relative to the single particle band gap defines the band gap renormalization due to many body effects. Extensive theoretical and experimental studies in the context of electron-hole droplet formation imply that the band renormalization is mainly a function of the carrier density but insensitive to the temperature/8,12/. Hence the plasma density can be determined from the band renormalization independently from the emission line shape.

For the low temperature EHD experiments the values of the band renormalization obtained from the energetic position of the emission are in good agreement with those calculated from the carrier density.

We have evaluated luminescence spectra in the temperature range between 30K and 300K. From the lineshape fits we obtain a plasma density that is almost independent of the excitation intensity but which increases approximately linearly with temperature. A similar behaviour has been observed for low temperature EHP in a variety of semiconductors/13/.

In Fig. 2 we depict the experimental values of the band renormalization extracted from the line position (triangles) together with those expected theoretically from the carrier densities derived from the fit (squares)/14/. In the entire region investigated the theoretical values of Eg are significantly smaller (discrepancy 10meV - 20meV) than the experimental ones. In the light of the good agreement of experiment and theory for the electron-hole droplets this disagreement is unexpected. In the following we shall demonstrate that this discrepancy is due to the unjustified use of a quasi-equilibrium theory for the description of the plasma.

IV - Drift Effects

In the previous theoretical considerations a homogeneous and stationary plasma state has been assumed. However the plasma is created at the surface of the sample and due to the high gradients of carrier density and temperature it will expand rapidly into the bulk. The fast drift leads to a net plasma momentum and therefore alters the distribution functions. In a first approximation we assume that the Fermi spheres are rigidly shifted in momentum space by average drift vectors \( \mathbf{k}_D \):

\[
f = \frac{1}{1+\exp(-\frac{\hbar^2}{2mk_B T}((\mathbf{k}-\mathbf{k}_D)^2-k_F^2))}
\]

As the luminescence is determined mainly by the distribution functions we expect significant changes for drift vectors comparable to the Fermi vectors.
We have reevaluated the luminescence spectra including the drifted Fermi distribution under the assumption that the density is determined most reliably from the line position. This means that $E_g$ is independent of $kD$ and a function of the density only. This is consistent with the very weak $k$-dependence of the theoretical band renormalization.

As shown in Fig. 3 the disagreement between experimental (triangles) and theoretical values (spheres) of $E_g$ is significantly decreased by including the carrier drift in the evaluation of the spectra. We obtain typical values of 4·$10^6$ cm/s for the plasma expansion velocity in good agreement with an independent measurement based on EHP time constants /15/.

An example of a line shape fit including the plasma drift is shown in Fig. 4. As the carriers are distributed over a wider energetic range if drift is included line shape fits with drifted Fermi functions yield significantly lower densities and slightly reduced temperatures compared to fits assuming quasi equilibrium. Note that the average electronic temperature derived from the fit does not exceed the bath temperature by more than 40K even during the excitation pulse.

V - Discussion

Three important results can be derived from the present study of EHP in Si excited to the onset of ns-PLA:

- The average plasma densities obtained for pulse energies within a factor of four below the single pulse annealing threshold are of the order of $10^{19}$ cm$^{-3}$ (see Fig. 5). These densities are approximately two orders of magnitude lower than necessary for the onset of plasma driven instabilities in the phonon system (non-thermal melting)/16/ and also much lower than required for the screening of the electron-phonon coupling/17/.
The plasma expands with drift velocities of about $4 \times 10^6 \text{cm/s}$, i.e. the carrier pairs traverse through $40 \mu\text{m}$ within $1 \text{ns}$. This means that even during a $10\text{ps}$ pulse the plasma occupies a volume about twice as large as the one excited by the laser. Hence in $\text{ns}$ as well as in $\text{ps}$ laser annealing experiments roughly $50\%$ of the excitation energy (for $2E_g$ excitation) leave the excited region. Furthermore the fast plasma drift suggests that the density reduction by drift is comparable to density losses by Auger recombination.

The small difference between the lattice and the electronic temperature shows clearly that on a $\text{ns}$ time scale the electron-hole pairs have transferred their excess energy ($\gtrsim 1 \text{eV}$) to the lattice. We find no evidence of a hot plasma persisting several nanoseconds.

In summary our study emphasizes that the plasma created by high surface excitation must not be treated by equilibrium thermodynamics but that its non-equilibrium nature has to be considered.

References

10. Note that the unstructured luminescence bands (see Fig.1) are the sum of $T0$, $L0$, and $TA$ phonon replica of the transition. To take into account the finite lifetime of the final states we use a formula proposed by Landsberg, which leads to a broadening at the low energy side of the emission line (see Ref.8).
14. The reduction of the single particle band gap due to temperature effects has been corrected according to Ref.11.